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Thermodynamic constraints on H_2 production during mantle wedge alteration: Clues from serpentinite to listvenite sequences

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Fluid-rock interactions can produce H_2 from the oxidation of iron in rocks coupled with the reduction of water to H_2. The mantle wedge, where iron-bearing peridotites react with fluids mobilized from the underlying subducting plate, can be a setting for significant natural H_2 production. Subduction-mobilized fluids transform the overlying mantle peridotites into various volatile-rich rock types, including serpentinites, soapstone (talc-magnesite rock), and listvenite (magnesite-quartz rock). The formation of these different rock types depends on the carbon content of the mobilized fluids. The dissolved CO_2 content in the reacting fluids influences the amount of H_2 produced during fluid-rock interactions. Specifically, CO_2-rich fluids favor siderite formation, which traps iron in the ferrous state, lowering opportunities for iron oxidation and H_2 production. Additionally, fluids with higher silica activities, near quartz saturation, form during interactions between peridotites and CO_2-rich fluids, thereby lowering the potential for H_2 production.

In this study, we used the DEW model and EQ3/6 software to compare and quantify H_2 production during interactions of mantle wedge peridotites with fluids containing varying levels of CO_2. Fluids sourced from subducted siliciclastic sedimentary rocks are enriched in dissolved CO_2 (~40,000 ppm C) due to devolatilization reactions that occur during calc-silicate metamorphism. These fluids contribute to the formation of listvenite and soapstone lithologies at high fluid-to-mantle rock ratios. However, H_2 production in this scenario is limited. Significant H_2 production (up to 400 millimole H_2/kg of rock reacted) occurs only when the fluid-to-mantle rock mass ratio drops below 2, allowing for serpentinization.

In contrast, fluids mobilized from limestone-rich sediments are influenced by the solubility of $CaCO_3$ minerals, yielding fluids with low levels of dissolved CO_2 (~1,000 ppm C). Listvenite does not form when these low-CO_2 fluids react with the mantle. Instead, a sequence of rock types—pyroxenite, amphibolite, talc schist, and serpentinite—develops. The lower CO_2 and silica activity of these fluids enhances H_2 production (100 to 500 millimole H_2/kg of rock reacted) across all fluid-to-rock mass ratios. In summary, H_2 production in subduction zones is highly dependent on the type of mantle metasomatism enabled by the composition of subduction-mobilized fluids.